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CITATION:

Nakano, Huzio. Hierarchical Variational Principles of Irreversible Processes in Dynamical and Thermal Disturbance. 数理解析研究所講究録 1997, 1013: 131-141

ISSUE DATE:

1997-09

URL:

http://hdl.handle.net/2433/61565

数理解析研究所講究録
1013巻 1997年 131-141



# Hierarchical Variational Principles of Irreversible Processes in Dynamical and Thermal Disturbance

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Quantum variational principles of irreversible processes in the theory of linear response which have been developed for the electric conduction by the present author and his coworker are generalized to the transport phenomena in thermal disturbance. The princple is also presented on the quantum fluctuation in the aged system, manifesting the fluctuation-dissipation law. The primary version is concerned with the dynamical stage and independent of irreversibility. By means of contractions of informations, the principle is converted into the more coarse grained one, which is thus concerned with irreversibility. In the first step the conversion takes place from the dynamical to kinetic stages and in the second from the kinetic to thermohydrodynamical stages.

## §1. Introduction

We have developed the variational principle of irreversible processes in the linear response theory, taking the electrical conduction in solids as a typical example.[1)2)3)4)] In the present paper we generalize the principle to the transport in the system driven by the thermal disturbance based on the theory due to Nakajima[5)]. He generalized the local equilibrium distribution function to the local equilibrium density matrix as the standard from which the state of the molecular state deviate. We also apply the principle to the theory of quantum-mechanical fluctuation in the aged system[6)] which generalized Onsager's stochastic thermodynamical theory giving the well-known reciprocity relation. The principles thus obtained in the deepest microscopic level of information is reduced to the more coarse grained level by contracting the informations in the density matrix of the system in the same way as in the our previous papers mentioned above. The contraction is made in two steps successively and as a result the variational principles at the levels of kinetic and thermohydrodynamical stages is derived.

In §2 we revise the previous variational principle on the electric couduction due to the applied electric field based on the von-Neumann eqution, by replacing the density matrix of the grand

canonical form for equilibrium with that for the local equilibrium.[6] Thus the variational principle is generalized to the thermally as well as dynmaiclly driven systems.

In §3 we discuss the thermal fluctuation in the aged system on the basis of the theory due to Nakajima et al.[7][8] and propose the variational principle of the same form as in the transport phenomena, manifesting the fluctuation-dissipation law. In §4, applying the information contractions, we obtain a hierachy of variational principles depicting the various dynamical stages of the system.

## §2. Formulation for the system in thermal distsurbance

The density matrix $\rho(t)$ for the system exposed to an electric field $\mathbf{E}(t)$ obeys a von-Neumann equation

$$i\hbar\partial\rho/\partial t=[H-\mathbf{P}\cdot\mathbf{E}(t),\rho] \tag{1}$$

where $\mathbf{P}$ represents the polarization operator of the system. We assume that the system is maintained at local equilibrium in thermal contact with the surrounding heat bath with temperatures varying in space, with transport phenomena being accompanied.

We can write he density matrix for this system as

$$\rho(t)=\rho_L+\rho_1(t) \tag{2}$$

in terms of the local equilibrium part

$$\rho_L=K\exp\left[-\int\{\beta_L(\mathbf{r})u(\mathbf{r})+\xi_L(\mathbf{r})n(\mathbf{r})\}d^3\mathbf{r}\right] \tag{3}$$

and the deviation $\rho_1(t)$. In (3) K is a nomalization constant, and $\beta_L(\mathbf{r})$ and $\xi_L(\mathbf{r})$ are local values of the inverse temperature and of the chemical potential divided by temperature, respectively. They are thermodynamically conjugate to the energy density $u(\mathbf{r})$ and the number density $n(\mathbf{r})$, respectively. Integrations of these densities give

$$H=\int u(\mathbf{r})\}d^3\mathbf{r}, \quad N=\int n(\mathbf{r})\}d^3\mathbf{r}, \tag{4}$$

where H and N are the Hamiltonian and the total number of electrons, respectively. The local parameters $\beta_L(\mathbf{r})$ and $\xi_L(\mathbf{r})$ are determined from the local averages $u_L(\mathbf{r})$ and $n_L(\mathbf{r})$ of energy and number densities with respect to the local as well as total equilibrium density matrices as

$$u_L(r)=\int \mathrm{Tr}[\rho(t)u(r)]d^3r=\int \mathrm{Tr}[\rho_L u(r)]d^3r, \tag{5}$$

$$n_L(r)=\int \mathrm{Tr}[\rho(t)n(r)]d^3r=\int \mathrm{Tr}[\rho_L n(r)]d^3r. \tag{6}$$

respectively.

Dividing the local parameters as

$$\beta_L(r)=\beta+\beta'(r), \quad \xi_L(r)=\xi+\xi'(r) \tag{7}$$

into the uniform values $\beta$ and $\xi$ and small deviations $\beta'$ and $\xi'$ which are sowly-varying with space and time, we expand (3) as

$$\rho_L(t)=\rho_C[1-\beta^{-1}\int d^3r\int_0^\beta e^{\lambda H}(\beta'u+\xi'n)e^{-\lambda H}d\lambda] \tag{8}$$

into a power series of $\beta'$ and $\xi'$ up to the first order, where

$$\rho_C=K\exp(-\beta H-\xi N) \tag{9}$$

is the density matrix for the system at thermal equilibrium. The integration with sapce in (8) extends all over the system. Substituting (8) into the von-Neumann equation (1), we obatin

$$\partial\rho_1/\partial t+i[H,\rho_1]/\hbar$$

$$=\int_0^\beta d\lambda\int d^3r e^{\lambda H}\left\{j\cdot E(t)-\beta^{-1}(\dot\beta'u+\dot\xi'n+\beta'\dot u+\xi'\dot n)\right\}e^{-\lambda H} \tag{10}$$

in the linear approximation with respect to the external disturbance. Here, in contrast to the fact that $\dot\beta'$ and $\dot\xi'$ simply denote the derivatives of $\beta'$ and $\xi'$ with time, respectively, $\dot u$ and $\dot n$ stand for the changes with time according to the Heisenberg equation of motion and obey the equation of continuity, so that

$$\dot n=i[H,n]/\hbar=-\nabla\cdot j/e, \quad \dot u=i[H,u]/\hbar=-\nabla\cdot w, \tag{11}$$

where $j$ and $w$ denote the electric current and energy current densities, respectively. In conformity of the left-hand side with the right-hand side in (10), we put

$$\rho_1=\int_0^\beta d\lambda\int d^3r e^{\lambda H}\Phi e^{-\lambda H}. \tag{12}$$

in (10) and perform partial integrations with space coordinates to get

$$\partial\Phi/\partial t+i[H,\Phi]/\hbar=\Pi+\beta^{-1}(\dot\beta'u+\dot\xi'n) \tag{13}$$

for the deviation operator $\Phi$. Here we have defined

$$\Pi = j\cdot X_1 + q\cdot X_2 = j\cdot X_1' + w\cdot X_2 \tag{14}$$

in terms of $j$ and $w$ or the heat flow $q$ given by

$$q = w - \mu j/e, \tag{15}$$

and their thermodynamically conjugate forces defined, respectively, as

$$X_1 = E - e^{-1}\nabla\mu, \quad X_2 = -T^{-1}\nabla T, \quad X_1' = E - e^{-1}T\nabla(\mu/T). \tag{16}$$

When $X_1$ and $X_2$ as well as $\beta'$ and $\xi'$ depend on time as

$$X_1(t) = X_1 e^{st}, \quad X_2(t) = X_2 e^{st}, \quad X_1'(t) = X_1' e^{st}, \quad (t<0) \tag{17a}$$

on the one hand and

$$X_1(t) = X_1 e^{-st}, \quad X_2(t) = X_2 e^{-st}, \quad X_1'(t) = X_1' e^{-st}, \quad (t\geq 0) \tag{17b}$$

on the other hand, we assume

$$\Phi(t) = \Phi^{(+)} e^{-st} \tag{18a}$$

and

$$\Phi(t) = \Phi^{(-)} e^{st}, \tag{18b}$$

respectively, where s will tends to plus zero in the following. Substituting (17a) and (17a) and next (18b) and (19b) into (14), we get

$$L_s \Phi^{(+)} = \Pi, \tag{19a}$$

$$L_{-s} \Phi^{(-)} = \Pi, \tag{19b}$$

respectively, where no contribution comes from the terms other than $\Pi$ in the integrand on the right-hand side of (14) as $s\to+0$. A superoperator $L_s$ acting on an operator $\Phi$ is defined by

$$L_s \Phi = s\Phi + i[H,\Phi]/\hbar, \tag{20}$$

and satifies

$$(\Phi, L_s \Psi) = -(\Psi, L_{-s}\Phi) \tag{21}$$

with respect to the inner product which is been defined between a pair of operators $\Phi$ and $\Psi$ as

$$(\Phi,\Psi) = (\Psi,\Phi) = \int_0^\beta \mathrm{Tr}(\rho_C e^{\lambda H}\Phi e^{-\lambda H}\Psi)\,d\lambda. \tag{22}$$

The variational principle is thus presented as follows. Let us make the functional

$$W(\Phi^{(+)},\Phi^{(-)})=(\Phi^{(+)}-\Phi^{(-)},j\cdot X_1+q\cdot X_2)+(\Phi^{(-)},L_s\Phi^{(+)}) \quad (23)$$

stationary with respect to $\Phi^{(+)}$ and $\Phi^{(-)}$ in the limit $s\to 0$. Thus the solutions satisfy (19a) and (19b), respectively, and are written

$$\Phi^{(\pm)}=\Phi_1{}^{(\pm)}\cdot X_1+\Phi_2{}^{(\pm)}\cdot X_2 \quad (24)$$

in terms of the vector operators $\Phi_1{}^{(\pm)}$ and $\Phi_2{}^{(\pm)}$ satisfying

$$L_s\Phi_i{}^{(+)}=j_i, \qquad (i=1,2) \quad (25a)$$

$$L_{-s}\Phi_j{}^{(-)}=j_j. \qquad (j=1,2) \quad (25b)$$

respectively, where we have redefined the flows as

$$j_1=j,\ j_2=q, \quad (26)$$

in conformity with the forces $X_1$ and $X_2$. We can adopt $w$ as $j_2$ in place of $q$, by replacing $X_1$ with $X_1'$ in (24).

A pair of flows are coupled to each other in the functional

$$W(\Phi_i{}^{(+)},\Phi_j{}^{(-)})=(\Phi_i{}^{(+)},j_j)-(\Phi_j{}^{(-)},j_i)+(\Phi_j{}^{(-)},L_s\Phi_i{}^{(+)}), \quad (27)$$

which is made stationary by the solutions of (25a) and (25b). The stationary value gives the transport coefficent

$$L_{ij}=(\Phi_i{}^{(+)},j_j)=(\Phi_j{}^{(+)},L_{-s}\Phi_i{}^{(-)})=-(\Phi_j{}^{(-)},j_i)=-(\Phi_i{}^{(-)},L_s\Phi_j{}^{(+)}) \quad (28)$$

in terms of the solutions of (25a) and (25b).

In discussing the time reversal symmetry, $\bar{Q}$ denotes the time reversal of an operator Q. In the Schrödinger representation, the time reversal is performed by taking the complex conjugate: e.g., $\bar{x}=x$ and $\bar{p}=-p$. The Hamiltonian $H(\mathbf{H})$ of the system which is exposed to a magnetic field $\mathbf{H}$ is transformed as

$$\bar{H}(\mathbf{H})=H(-\mathbf{H}). \quad (29)$$

In the absence of magnetic field, to which we confine ourselves, $H\equiv H(0)$ is real and invaraiant with time inversion: $\bar{H}=H$. Since $H$ is real, if $|n\rangle$ is an eigenket, then its complex conjugate $|\bar{n}\rangle$ which denotes the time reversed state of $|n\rangle$ is also an eigenket with the same eigenvalue $E_n$ as $|n\rangle$: that is,

$$H|n\rangle=E_n|n\rangle, \quad H|\bar{n}\rangle=E_n|\bar{n}\rangle. \quad (30)$$

We also have

$$\langle m|n\rangle=\langle\bar{n}|\bar{m}\rangle, \quad \langle m|Q|n\rangle=\langle\bar{n}|\bar{Q}|\bar{m}\rangle, \tag{31}$$

where Q is hermitian. Otherwise, $\bar{Q}$ should be read the Hermite conjugate $\bar{Q}^{\dagger}$ of $\bar{Q}$.

Concerning the matrix element on the left-hand side of (20a) for the Hermitian operator $\Phi^{(+)}$, we have the relation

$$\langle m|L_s\Phi^{(+)}|n\rangle=-\langle\bar{n}|L_{-s}\overline{\Phi^{(+)}}|\bar{m}\rangle, \tag{32}$$

where $\omega_{mn}\equiv(E_m-E_n)/\hbar$. Taking $\Pi$ as Q in (31), we get

$$\langle m|\Pi|n\rangle=-\langle\bar{n}|\Pi|\bar{m}\rangle, \tag{33}$$

since $\bar{\Pi}=-\Pi$. Taking into consideration (20a), (32) and (33), we find that $\overline{\Phi^{(+)}}$ satisfies (20b). Thus we can assume

$$\overline{\Phi^{(+)}} = \Phi^{(-)}, \tag{34}$$

and simply write $\Phi^{(+)}$ as $\Phi$ and $\Phi^{(-)}$ as $\bar{\Phi}(\bar{\Phi}_i)$. Defining

$$D_{m,n}\equiv(\rho_n-\rho_m)/(E_m-E_n), \tag{35}$$

we can demonstrate

$$(\Phi_j^{(-)},L_s\Phi_i^{(+)})=\sum_m\sum_n D_{m,n}\langle m|\Phi_j^{(-)}|n\rangle\langle n|L_s\Phi_i^{(+)}|m\rangle$$

$$\sum_{\bar{m}}\sum_{\bar{n}}D_{\bar{m},\bar{n}}\langle m|\Phi_i^{(-)}|n\rangle\langle n|L_s\Phi_j^{(+)}|m\rangle=(\Phi_i^{(-)},L_s\Phi_j^{(+)}), \tag{36}$$

based on the relations (32) and (21) and the identities

$$\rho_{\bar{n}}=\rho_n, \quad D_{\bar{m},\bar{n}}=D_{m,n} \tag{37}$$

due to the degeneracy (32) in the energy eigenvalues. Taking into account (36) in (28) leads to Onsager's reciprocity relation[7],

$$(L_{ij})_{\mu\nu}=(L_{ji})_{\nu\mu}. \tag{38}$$

Substituting the solution of (25a)/(25b) into (28), we obtain

$$(L_{ij})_{\mu\nu}=\int_0^{\infty}dt(j_{i\mu}(t),j_{j\nu}) \tag{39}$$

in the limit $s\rightarrow+0$, where $j_{i\mu}$ and $j_{j\nu}$ denote the $\mu$-component of $j_i$ and the $\nu$-component of $j_j$, respectively. When the system is isotropic or has a cubic symmetry, (38) is rewritten as

$$(L_{ij})_{\mu\nu}=L_{ij}\delta_{\nu\mu}, \quad L_{ij}=L_{ji} \quad (\mu,\nu=x,y \text{ or } z) \tag{40}$$

in terms of the Kronecker delta symbol $\delta_{\nu\mu}$ and

$$L_{ij}=\int_0^\infty dt\int_0^\beta d\lambda\ \mathrm{Tr}\{\rho_C j_i(t-i\hbar\lambda)j_j\}, \tag{41}$$

where $j_{i\mu}$ and $j_{j\mu}$ have been abbreviated as $j_i$ and $j_j$, respectively.

When the system is exposed to an external magnetic field $\mathbf{H}$, the Schrödinger equations (30) are replaced with

$$H(\mathbf{H})|n\rangle=E_n|n\rangle,\quad H(-\mathbf{H})|\bar{n}\rangle=E_n|\bar{n}\rangle, \tag{42}$$

based on (29). Thus the field dependence of (37) is written as

$$\rho_{\bar{n}}(\mathbf{H})=\rho_n(-\mathbf{H}).\quad D_{\bar{m},\bar{n}}(\mathbf{H})=D_{m,n}(-\mathbf{H}), \tag{43}$$

wherefrom we obtain

$$(L_{ij}(\mathbf{H}))_{\mu\nu}=(L_{ji}(-\mathbf{H}))_{\nu\mu}\quad(\mu,\nu=x,y,z) \tag{44}$$

for the magnetic field-dependent coefficient $L_{ij}(\mathbf{H})$.

## §3. Fluctuating system

Assuming that the local temperature and chemical potential fluctuate slowly in the aged system where much faster fluctuations in molecular level occur, we write the density matrix of this aged system as[7]

$$\rho(t)=K\exp\left[-\int\{\beta_L(\mathbf{r})u(\mathbf{r})+\xi_L(\mathbf{r})n(\mathbf{r})+\Phi(t)\}d^3\mathbf{r}\right], \tag{45}$$

where $\Phi(t)$ denotes the rapid molecular fluctuation. We divide $\beta_L(\mathbf{r})$ and $\xi_L(\mathbf{r})$ as in (7), where $\beta$ and $\xi$ denote the equilibrium value and $\beta'$ and $\xi'$ are not deviations driven externally but internal thermal fluctuations. Substituting (7) into (45), we regard $\beta'$ and $\xi'$ as well as $\Phi$ to be small and expand into a power series of these, we get

$$\rho(t)=\rho_C[1-\beta^{-1}\int d^3\mathbf{r}\int_0^\beta e^{\lambda H}(\beta\Phi+\beta'u+\xi'n)e^{-\lambda H}d\lambda], \tag{46}$$

where $\rho_C$ denotes the equilibrium density matrix (9).

Substituting (46) into the von-Neumann equation

$$i\hbar\partial\rho/\partial t=[H,\rho], \tag{47}$$

for the isolated system with the Hamiltonian H, we get (13) again, where we have to notice the physical meaning. What we are concerned with here is the fluctuations in the aged sytem, in contrast with

the dissipations in the system driven externally in the last section. By taking into consideration that the regression of the thermal fluctuations are much slower than that of the fluctuations in molecular level, $\beta^{-1}(\dot{\beta}'u+\dot{\xi}'n)$ can be neglected in comparison with $\Pi$ on the right-hand side of (13). Thus we obtain

$$\frac{\partial\Phi}{\partial t}+\frac{i}{\hbar}[H,\Phi]=j\cdot X_1+q\cdot X_2=j\cdot X_1'+w\cdot X_2\,. \tag{48}$$

If we assume the regression of $\Phi(t)$ as well as $X_1(t)$ and $X_2(t)$ in the form (19a) and (18a), we obtain (20a). Assuming (19b) and (18b) instead, we get (20b). Thus the variational principle concerning (23) applied to the dissipation phenomena in the driven system is here applicable to the thermal fluctuation in the aged system, so that it is needless to repeat the argument therein. This formal equivalence manifests the fluctuation-dissipation law in the form of the variational principle.

The variational principle concerning (24) can be rewritten in the time space in the form

$$\int_0^T dt\int d^3r[(\Phi(t-T),\Pi(t))-(\Phi(t),\Pi(t-T))-(\Phi(t-T),\partial\Phi(t)/\partial t+L\Phi(t))] \tag{49}$$

where

$$\Pi(t)=j\cdot X_1(t)+q\cdot X_2(t)=j\cdot X_1'(t)+w\cdot X_2(t). \tag{50}$$

Let us make (49) stationary w. r. t. $\Phi(t)$, we get the von-Neumann equation (48) in the time range $-T<t<T$. In the limit $T\to\infty$, (49) is rewritten as (24) in terms of the Lapalace transform

$$\Phi^{(+)}=\int_0^\infty e^{-st}\Phi(t)dt,\qquad \Phi^{(-)}=\int_{-\infty}^0 e^{st}\Phi(t)dt. \tag{51}$$

§4. Informational contractions and hierarchy

The principle developed in the foregoing sections is concerned with the dynmaicl or initial-mixing stage, where the von Neumann equation governs the principle. The time reversal symmetry satisfied by that principle is broken by eliminating the even component with respect to time reversal which is found to be irrerevant. Thus the princple is transformed into the kinetic stage inplicating irreversibility. The density operators introduced by (19a) and (19b) are divided as

$$\Phi^{(+)}=\Phi'+\Phi'',\quad \Phi^{(-)}=-\Phi'+\Phi'', \tag{52}$$

where $\Phi'$ and $\Phi''$ are odd and even components with respect to time reversal, respectively. That is,

$$\overline{\Phi'}=-\Phi', \quad \overline{\Phi''}=\Phi''. \tag{53}$$

The even component $\Phi''$ is irrelevant in the variation of (24), because it is decoupled with the term of external disturbance. By making (24) stationary as to $\Phi''$, this is eliminated irrespective of the external term and expressed in terms of $\Phi'$ as a result. Substituting this expression into (24), we get

$$W(\Phi')=2(\Phi',\Pi)-(\Phi',\mathrm{L}\Phi'), \tag{54}$$

which is maximized w. r. t. $\Phi'$. The superoperator $\mathrm{L}$ is defined by

$$\mathrm{L}\Phi'=([H,[H,\Phi]]+\hbar^2 s^2\Phi)/(\hbar^2 s) \tag{55}$$

and satisfies the relations

$$(\Phi,\mathrm{L}\Psi)=(\Psi,\mathrm{L}\Phi), \tag{56}$$

$$(\Phi,\mathrm{L}\Phi)\geq 0. \tag{57}$$

The maximization of (61) demands

$$\mathrm{L}\Phi'=\Pi \tag{58}$$

of $\Phi'$, where the maximum is the observed value $(\Phi',\Pi)$ of $\Pi$ and a traxpot coefficient in a special case. This variational principle is formally the same as the the Umeda-Kohler-Sondheimer(UKS) principle which was originally presented on the Boltzmann equation for the conduction electrons in solids[8] and for molecules in gases[9]. It is noticed that eq. (58) as well as the Boltzmann equation are self-adjoint in contrast with the von-Neumann equation (20a) whose adjoint is given by (20b).

For brevity we here confine ourselves to the isotropic or cubic system and delete the suffix for the orthogonal component. The solution of (58) can be written as

$$\Phi'= X_1\Phi_1+X_2\Phi_2, \tag{59}$$

which maximizes (54) if $\Phi_n$ satisfies a Boltzamann type equation

$$\mathrm{L}\Phi_n=j_n \quad (n=1,2). \tag{60}$$

The maximum is written as

$$W(\Phi')=L_{11}X_1^2+2L_{12}X_1X_{21}+L_{22}X_2^2, \tag{61}$$

in terms of the matric transport coefficient

$$L_{mn}=(\Phi_m, j_n)=(\Phi_m, \mathrm{L}\Phi_n)=\int_0^\infty (j_m, e^{i\mathrm{L}t-st} j_n)dt \quad (s\to\infty). \tag{62}$$

The solution of (60) maximizes

$$W(\Phi_n)=2(\Phi_n, j_n)-(\Phi_n, \mathrm{L}\Phi_n) \qquad (n=1,2), \tag{63}$$

where the maximum is equal to $L_{nn}$. Substituting (66) into the averages of the flow $J_m=(\Phi', j_m)$, we obtain

$$J_m=\sum_n L_{mn}X_n \qquad (m=1,2), \tag{64}$$

where $L_{mn}$ defined as (62) satisfies Onsager's reciprocity[7]

$$L_{mn}=L_{nm} \qquad (m,n=1,2). \tag{65}$$

Substituting (59) and (15) into (54) and taking into account (62) and (64), we obtain

$$W(J)=\sum_m\sum_n [2J_nX_n-(L^{-1})_{mn}J_mJ_n] \tag{66}$$

in terms of the inverse $L^{-1}$ to L. Asuming that $\{L_{mn}\}$ is known in (66) for the system concerned, we obtain a variational principle of Onsager's type[7], where (66) is maximized w. r. t. $\{J_n\}$.

§4.Conclusion

The quantum variational principle of irreversible processes based on the von Neumann equation is generalized to the transport due to the thermal disturbance, where a pair of solution are coupled with each other similarly to the incoming and outgoing waves in the scattering theeory.[9] It describes the dynamical stage, having no concern with irreversibility, where the pair of solutions of the von-Neumann equation are adjoint to each other, in contrast with the self-adjoint Boltzmann equation.

By contracting the even component $\Phi''$ of the density matrix, we enter in the kinetic regime which concerns the irreversibility, and get the UKS principle on the odd component as a maximum problem. By means of a further contraction, we enter in the thermohydrodynamical regime, where we gain the Onsager type variational principle on the flow quantities describing the transport.

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